

PATENT ABSTRACTS OF JAPAN

(11)Publication number : 2003-160316

(43)Date of publication of application : 03.06.2003

(51)Int.Cl. C01B 31/02

(21)Application number : 2001-357006 (71)Applicant : MITSUBISHI CHEMICALS CORP

(22)Date of filing : 22.11.2001 (72)Inventor : TAKEHARA HIROAKI

YAMAMOTO TAKAHARU

(54) METHOD AND APPARATUS FOR PRODUCING FULLERENES

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a method and an apparatus able to produce economically a fullerene with mass production.

SOLUTION: Fullerenes are produced by combustion and/or pyrolysis of a carbon containing compound in a combustion furnace. An oxygen containing gas fed for a combustion reaction is preheated.

LEGAL STATUS

[Date of request for examination]

[Date of sending the examiner's decision of rejection]

[Kind of final disposal of application other than the examiner's decision of rejection or application converted registration]

[Date of final disposal for application]

[Patent number]

[Date of registration]

[Number of appeal against examiner's decision of rejection]

[Date of requesting appeal against examiner's decision of rejection]

BEST AVAILABLE COPY

[Date of extinction of right]

Copyright (C); 1998,2003 Japan Patent Office

(19)日本国特許庁 (JP)

(12) 公開特許公報 (A)

(11)特許出版公開番号

特開2003-160316

(P2003-160316A)

(43)公開日 平成15年6月3日(2003.6.3)

(51)Int.Cl.
C01B 31/02識別記号
101F I
C01B 31/02テ-テ-ト(参考)
101F 4G046

審査請求 未請求 請求項の数11 OL (全 5 頁)

(21)出願番号 特願2001-357006(P2001-357006)

(71)出願人 000005968

(22)出願日 平成13年11月22日(2001.11.22)

三菱化学株式会社

東京都千代田区丸の内二丁目5番2号

(72)発明者 武原 弘明

福岡県北九州市八幡西区星崎城石1番1号

三菱化学株式会社内

(72)発明者 山本 隆晴

福岡県北九州市八幡西区星崎城石1番1号

三菱化学株式会社内

(74)代理人 100103997

弁理士 長谷川 曜司

Pターム(参考) 4C046 C001 C002 C005 C002 C003

C009

(54)【発明の名称】 フラーレン類の製造方法およびフラーレン類の製造装置

(57)【要約】

【課題】 経済的かつ大量生産可能なフラーレン製造方法および製造装置を提供する。

【解決する手段】 燃焼炉内において炭素含有化合物を燃焼及び/または熱分解させてフラーレン類を生成するフラーレン類の製造方法であって、燃焼反応のために供給する酸素含有ガスの温度を予熱する事を特徴とするフラーレンの製造方法。

(2)

特開2003-160316

1

2

【特許請求の範囲】

【請求項1】 燃焼炉内において炭素含有化合物を燃焼及び/または熱分解させてフラー-レン類を生成するフラー-レン類の製造方法であって、燃焼反応のために供給する酸素含有ガスの温度を予熱する事を特徴とするフラー-レン類の製造方法。

【請求項2】 予熱の温度が燃料である炭素含有化合物の自己着火温度以上である事を特徴とする請求項1に記載のフラー-レン類の製造方法。

【請求項3】 供給する酸素含有ガスが不活性ガスを含有することを特徴とする請求項1または2に記載のフラー-レン類の製造方法。

【請求項4】 供給する酸素含有ガスが空気である事を特徴とする請求項1乃至3のいずれかに記載のフラー-レン類の製造方法。

【請求項5】 反応炉内の圧力が大気圧未満である事を特徴とする請求項1乃至4のいずれかに記載のフラー-レン類の製造方法。

【請求項6】 燃焼炉内において炭素含有化合物を燃焼及び/または熱分解させてフラー-レン類を生成するフラー-レン類の製造方法であって、燃焼状態が高温空気燃焼であることを特徴とするフラー-レン類の製造方法。

【請求項7】 平均燃焼温度が1000°C以上である請求項1乃至6のいずれかに記載のフラー-レン類の製造方法。

【請求項8】 反応炉内の圧力が大気圧未満である、請求項1乃至7のいずれかに記載のフラー-レン類の製造方法。

【請求項9】 燃焼炉内において炭素含有化合物を燃焼及び/または熱分解させてフラー-レン類を生成するフラー-レン類の製造方法であって、燃料供給口と酸素含有ガス供給口とが各々独立に距離を隔てて反応炉の同一側に開口していることを特徴とするフラー-レン類の製造装置。

【請求項10】 酸素含有ガス供給口中に更に燃料供給口を有する請求項9に記載の装置。

【請求項11】 反応炉内に開口した酸素含有ガス供給口の形状が非円形である請求項9または10に記載の装置。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】 本発明は、フラー-レン類の製造方法及び製造装置に関する。

【0002】

【従来の技術】 フラー-レン類（以下、単にフラー-レンと言うことがある。）は、ダイヤモンド、黒鉛に次ぐ第三の炭素同素体の総称であり、 C_{60} 、 C_{70} などに代表されるようなら員環と6員環のネットワークで閉じた中空殻状の炭素分子である。フラー-レンの存在が最終的に確認されたのは比較的最近の1990年のことであり、

10

20

30

40

50

比較的新しい炭素材料であるが、その特殊な分子構造ゆえに特異的な物理的性質を示すことが認められ、例えば以下のような広範囲な分野に渡り、革新的な用途開発が急速に展開されつつある。

（1） 鋼鉄材料への応用：フラー-レンを前駆体として微細結晶粒子をもつ人工ダイヤモンドの結晶が可能なため、付加価値のある耐摩耗材料への利用が期待されている。

（2） 医薬品への応用： C_{60} 誘導体、光デバイスを用いることで抗癌剤、エイズ、骨粗鬆症・アルツハイマー治療薬、造影剤、ステント材料等の用途としての研究が進められている。

（3） 鋼伝導材料への応用：フラー-レン薄膜に金属カリウムをドープすると18Kという高い転移温度を持つ鋼伝導材料をつくり出すことができる事が発見され、多方面から注目を集めている。

（4） 半導体製造への応用：レジストに C_{60} を混ぜることでレジスト構造がより一層強化されることを利用し、次世代半導体製造への応用が期待されている。

【0003】 各種炭素素のフラー-レンの中でも C_{60} 、および C_{70} は比較的合成が容易であり、それゆえ今後の需要も爆発的に高まることが予想されている。現在知られているフラー-レンの製造方法としては以下に示す方法が挙げられる。

（1） レーザー蒸着法：希ガス中に置かれた炭素ターゲットに高エネルギー密度のパルスレーザーを照射し、炭素原子の蒸発により合成する方法。希ガスが流れれる石英管を電気炉の中に置き、グラファイト試料をその石英管の中に置く。ガスの流れの上流側からグラファイト試料にレーザーを照射し、蒸発させると電気炉出口付近の冷えた石英管の内壁に C_{60} や C_{70} などのフラー-レンを含む煤が付着する。ショット当たりの蒸発量がわずかであり、大量製造には向き。

（2） 抵抗加熱法：ヘリウムガスで満たされた真空の容器の中でグラファイト管を通過加熱し昇華させる方法。回路での電気抵抗ロスが大きいので大量製造に向き。

（3） アーク放電法：数十kPa中のヘリウムガス中で2本のグラファイト電極を軽く接触せたり、あるいは1~2mm程度離した状態でアーク放電を起こし、陽極の炭素を昇華させる方法。現在工場規模での大量製造に用いられている。

（4） 高周波誘導加熱法：抵抗加熱やアーク放電を使う代わりに、高周波誘導により原料グラファイトに過電流を流し、これを加熱・蒸発する方法。

（5） 燃焼法：ヘリウム等の不活性ガスと酸素との混合ガス中でベンゼン等の炭化水素原料を不完全燃焼させる方法。ベンゼン燃料の数%が煤となり、その10%程度がフラー-レンとなる点で製造効率は良くないが、複数の炉（フラー-レン等）を液体燃料等に使用可能なこ

(3)

特開2003-160316

3

と、製造装置が単純である点で、アーク台成法に対抗する大量生産法として注目されている。

(6) ナフタレン熱分解法：ナフタレンを約1000°Cで熱分解させる方法。

【0004】このように現在までにさまざまなフラー
レンの合成法が提案されているが、いずれの方法によ
ってもこれまでにフラーレンを安価に大量に製造する方法は
確立されていない。これらの方法のうち、最も安価で、
効率的な製造方法の一つと考えられるのは燃焼法であ
り、特表平6-507879号公報には、炭素含有物を
火炎中で燃焼させ凝縮物を収集することによるフラー
レンの製造方法が記載されている。フラーレンはフラー
レン等のすす状物質中に含まれて生成されるが、このすす
状物質中にフラーレンが含まれる割合をいかに高めるか
が大きな課題となっている。

【0005】上記、特表平6-507879号公報には
フラーレンの収率を向上させるために、火炎温度を上昇
させる事、またその手段として外部エネルギー源から火
炎にさらにエネルギーを供給する方法が述べられてい
る。好ましいエネルギー源としては、入力流の電気抵抗
加熱、マイクロウェーブ加熱、放電加熱及び向流加熱が
挙げられている。

【0006】また、一般的に、フラーレンの製造は、減
圧下で行われ、反応領域中に希釈剤を導入する場合もあ
る。これらの減圧度、希釈剤濃度は上記フラーレンの収
率に影響を及ぼす事が知られている。上記、特表平6-
507879号公報では、燃焼反応のための酸化剤として
純酸素が、また希釈剤としてはアルゴンが用いられて
いる。これは、フラーレンの収率を上げる効果があると
考えられる。

【0007】しかしながら、純酸素は専用のポンベもし
くは供給設備等を要し、特に、工業規模でフラーレンを
製造しようとする場合には、燃焼のために必要とする酸
素の量も大量になり、特別な酸素供給設備が必要で、結果
としてフラーレンの製造コストも高価となる。そこで、燃
焼法において製造コストを低減するために、燃焼
の酸化剤として空気を用いる事は容易に頼れるが、
純酸素に比べて酸素濃度が低いために火炎が安定しない
事や空素の割合が多いため燃焼温度が低くなる事、特に
減圧下での操作時に体積が増えノズルを通過する速度が
速くなる等の理由により実用化には至っていない。

【0008】一方、燃焼方法自体については、一般的な
工業用加熱炉において、通常の燃焼に比べて十分に熱発
生速度が低速な酸化発熱反応を伴い、平均熱流束を最大
熱流束に近づけてNO_xを抑制する燃焼方法として、い
わゆる高溫空氣燃焼法が知られている。工業用加熱炉に
おいてNO_xを抑制する別の燃焼方法として、炉内燃料
直接噴射法が知られている。詳しくは、燃焼用空気と燃
料を独立したノズルから炉内に噴射し、その噴出エネル
ギーによる自己排ガス再循環効果によって、周囲の燃焼

4

ガスを吸引し燃焼用空気の酸素濃度の低減と、燃焼時の
火炎温度の低下をもたらせる方法である。

【0009】また、上記の高溫空氣燃焼法と炉内直接噴
射法を組み合わせた燃焼方法も知られている。しかし、
いずれの公報にもフラーレンの製造方法および装置に關
する記載は全くない。フラーレンは次世代を担う新材
料、新素材として多方面から注目されており、フラー
レンを大量に且つ安価に、そして容易に製造する技術の開
発が望まれている。

【0010】

【発明が解決しようとする課題】本発明は前述した様な
事情に鑑みてなされたものであり、燃焼方法によるフラー
レンの製造において、燃焼炉中での燃焼を均一な燃焼
とし、フラーレンを大量に且つ安価に、そして容易に製
造する方法を提供することを目的とする。

【0011】

【課題を解決するための手段】本発明者らは、フラー
レンを大量に且つ安価に製造できる最適な燃焼方法を複々
検討した結果、燃焼に供給する酸化剤の温度を常温以上
に予熱し、かつ燃焼炉内の圧力を減圧することによ
って、フラーレンを安定的に生成できるとの知見を得た。

【0012】さらに酸化剤の温度を常温以上に予熱する
ことにより、燃焼温度の高溫化および火炎の安定化が容
易に図られるため、酸化剤として純酸素を用いすとも、
例えばアルゴン等の希ガスで希釈可能であるとの知見も
得た。また、この方法により酸化剤の酸素濃度が下げる
ため、純酸素のような高価なガスを用いすとも、例
えば空気のように入手容易な酸化剤にてフラーレンが生
成可能であるとの知見も得た。

【0013】一般的に燃焼法におけるフラーレンの収率
は、燃焼火炎温度が高いほど高くなる。火炎温度を高め
る方法としては、酸素の添加が知られているが、フラー
レンの製造の場合、酸化剤として純酸素を用いることも
あり、それ以上の酸素の添加は不可能である。ここで、
炉内に供給する酸化剤の温度を予熱することで容易に火
炎温度を高める事ができる。

【0014】更に、この温度を燃料炭化水素の自己着火
温度以上まで高めると、燃焼における反応炉内の温度
分布がより均一になり、フラーレンの生成をより効率的
に行う事ができる。また、この方法により燃焼反応が安
定化するため、純酸素に希釈剤を加え、酸素濃度を低下
させても安定的にフラーレンを生成する事ができる。

【0015】更に、この方法により、酸化剤として空気
を用いる事ができる。これにより、フラーレン製造にお
けるコストを大幅に低減する事ができる。空気を用いる
場合、空気中に窒素が含まれているため、排気ガス中に
NO_xが含まれ、その濃度は空気の予熱温度を高くする
ほど高くなる。このNO_x濃度上昇を抑制するため、供
給する空気の酸素濃度を低下させる方法、いわゆる高溫
空氣燃焼法や炉内に酸化剤と燃料をそれぞれ独立に距離

59

(4)

特開2003-160316

5

を隔てて導入するいわゆる炉内直接噴射法などが知られているが、これら方法をフラー・レン製造方法に応用し、かつ反応炉内の圧力減圧下にする事で、安価な空気を用いて安定的にフラー・レンを製造する事ができる。

【0016】

【発明の実施の形態】先ず、本発明に係るフラー・レンの製造装置および方法について説明する。図1は本発明に係るフラー・レン製造装置の一例の全体概略断面図、図2は酸化ガス導入用ノズルと燃料導入ノズルの配置説明図である。一般に燃料供給口(2)から燃料炭化水素を、酸素含有ガス供給口(3)から酸素含有ガスを供給し、これらを燃焼させることで高温の燃焼ガス流を反応炉の下流に向かって発生させる。酸素含有ガスとしては空気、酸素ガスまたはこれらにアルゴンガス等の不燃性ガスを任意の割合で混合したガスを使用することが出来、フラー・レンの収率という観点からは酸素ガスが好ましく、酸素含有ガスの入手のし易さ等の観点からは空気が好ましい。特に燃焼温度を上げるために、これらの酸素含有ガスは炉内に供給される前に予熱される。予熱の方法としては、熱交換器を使用した燃焼排ガスとの熱交換、いわゆるリジェネレーションバーナー等、公知のいかなる方法を用いても良い。この予熱の温度は常温以上であればいかなる温度でも良いが、フラー・レンの収率をあげるために極力高温度の方が好ましい。より好ましくは、燃料の自己着火温度以上である事が好ましい。この場合、一般的な金属製の熱交換器では、用いる金属の耐熱温度によって、酸化剤の最高予熱温度が決まってくるが、一般的に知られている耐熱金属を用いた場合、最高600°C程度が限界である。よって、自己着火温度以上に予熱しようとする場合は、レジエネレーションバーナー等、特殊な構造を持つバーナーを使用することが必要な場合もある。

【0017】燃焼反応に寄与する酸化剤の温度を炉内で予熱する方法として、酸化剤の炉内への供給流速を高速にすることで、炉内に燃焼排ガスの自己再循環流を形成させる方法をとっても良い。この場合、炉内へ供給する酸素含有ガスの流速は5m/s以上が好ましく、より好ましくは10m/s以上である。また、酸化剤供給口の形状を矩形にしたり、燃料ガスと酸化剤ガスとの交点の位置を工夫する事で、炉内において酸素含有ガスが少なくとも燃料と接触する前に、炉内排ガスを巻き込んで、酸素含有ガスの温度を燃料の自己着火温度以上とする事もできる。この場合は、さらに酸素含有ガス中の酸素濃度も着脱され低下するために、炉内の燃焼をいわゆる高温空気燃焼状態とすることもできる。このような燃焼状態では、炉内の温度はより均一で高温となるため、フラー・レンの収率をより高めることができる。

【0018】安定した高温空気燃焼を維持するため、後述する様に酸素含有ガス供給口に燃料供給口を設け、酸素含有ガスの一部を通炉燃焼することにより、酸素含

有ガスの温度を上げ且つ酸素濃度を低下させてもよい。燃料炭化水素及び原料炭化水素としては、水素、一酸化炭素、天然ガス、石油ガス等の燃料ガス、重油などの石油系液体燃料、クレオソート油などの石炭系液体燃料を使用することが出来る。中でもこれらを精製した芳香族系炭化水素を用いることが好ましく、特にベンゼンやトルエン等の芳香族系炭化水素が好ましい。原料の純度は高い方が好ましく、中でも芳香族系炭化水素を用いる際には純度が100%に近いほど良い。

【0019】またフラー・レンの収率を上げるために、燃焼も希ガス等を用いて着脱する事が好ましい。炉内の燃焼状態をいわゆる高温空気燃焼状態とするためには、公知のいかなる方法をとっても良い。図1に示すフラー・レン製造装置においては、燃料供給口(2)及び酸素含有ガス供給口(3)は、各自独立に反応炉の同一側に開口している。反応炉内に開口している各供給口の形状は任意であり、略円形、指円状、三角、四角状などの多角形状やひょうたん型などの不定形であってもよい。本発明者らの知見によれば、円形よりも、長円形や長方形の様に長径と短径を持つ形状の方が、酸素含有ガスの加热や着脱の速度がより速まる。従って、燃料供給口(2)としては、指円状や略円形が好ましく、酸素含有ガス供給口(3)としては、スリット状などの長方形形状が好ましく、これらを組み合わせるのが特に好ましい。

【0020】燃料供給口(2)と酸素含有ガス供給口(3)の配置は、各自独立に反応炉の同一側に開口していれば任意である。燃料の負荷やバーナー本数などの炉設計条件により、図2(A)～(E)に示す様いろいろな配置を採ることが出来るが、特に、図2(D)の様に、各自の供給口を反応炉の軸方向断面の中心を同心とする同一または同心円周上に、周方向に交互に配置するならば、炉内燃焼状態がより均一となるので好ましい。この際に、酸素ガス供給口(6)の形状が長径および短径を持つ様な場合には、長径から延びた直線が円の中心を通る様に配置するのが好ましい(図2(E)参照)。また、何れの供給口も、その開口端部が反応炉内の壁面と略同一平面上にあっても、突出していくてもよいが好ましくは略同一平面上がよい。

【0021】燃料供給口(2)及び酸素含有ガス供給口(3)から反応炉内に供給される燃料流および酸素含有ガス流は、各自の開口端部から、各供給口が配置されている炉壁面に対して任意の角度で供給してよいが、好ましくは略垂直となる様に、更には、供給される燃料および/または酸素含有ガスが開口端部から流の中心から略同心円状に拡散する様に供給するのが好ましい。

【0022】また、本発明の要件を満たす範囲においては、例えば酸素含有ガス供給口(3)中に更に燃料供給口(2)を設けてもよい。これは、炉の立ち上げ時など、炉内の温度が低温である場合、または、高温であっても炉内の燃焼温度やフラー・レンの物性などを制御した

50

(5)

特開2003-160316

8

7

い場合などに、この酸素含有ガス供給口(3)中に設置された燃料供給口(2)から燃料を供給し、局的に燃焼状態を変化させることにより、炉内の燃焼状態を制御し、より安定な操業を行うことが出来るからである。

【0023】反応炉内に供給される酸素含有ガス流および燃料流の流速は適宜選択すると共に反応炉内の温度変化などに応じて調整すればよい。図1に示すフーレン製造装置においては、炉中央部に更に燃料供給口(4)および酸素含有ガス供給口(5)を設けている。炉内の燃焼温度は重要で、少なくとも1000°C以上、中でも1400°C以上、更には1800°C以上、特に2000°C以上とするのが好ましい。炉内圧力は大気圧未満であることが好ましく、より好ましい範囲は10~300torrである。

*

* 【0024】燃料の希釈割濃度は、実質的に0~40モル%の範囲であり、また酸素含有ガスの希釈割濃度は0~90モル%の範囲で任意に調整できる。

【0025】

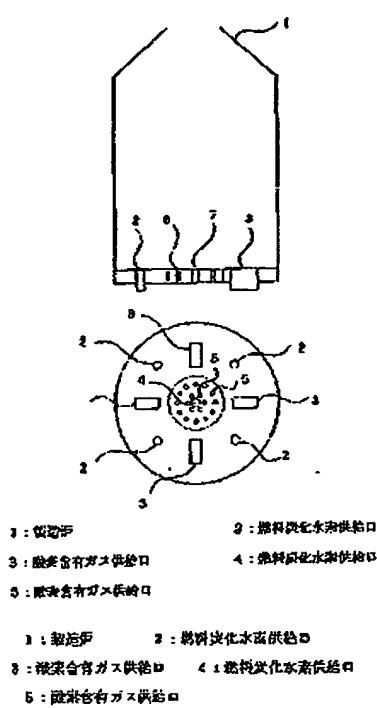
【発明の効果】本発明の製造方法によって、経済的な燃焼方法によるフーレンの大口径且つ安価な製造方法を提供することが出来る。

【図面の簡単な説明】

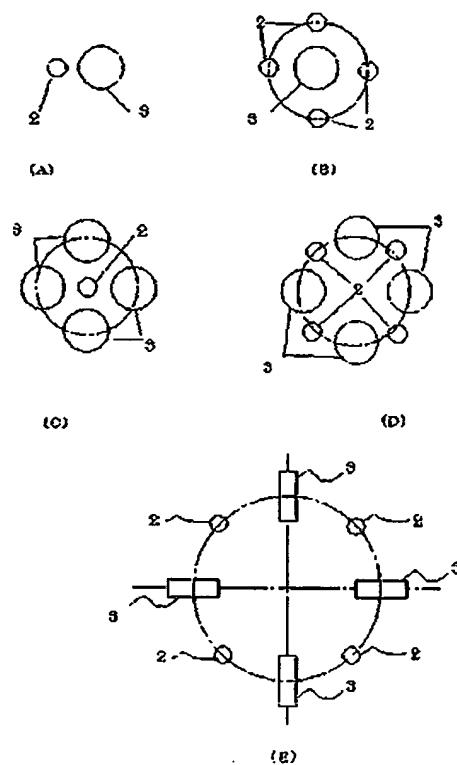
【図1】 図1は本発明に係るフーレン製造装置の一例の全体概略断面図、及び酸化ガス導入用ノズルと燃料導入ノズル、原料導入ノズル配置例の説明図である。

【図2】 図2は酸化ガス導入用ノズルと燃料導入ノズル、原料導入ノズルの配置説明図である。

【図1】



【図2】



* NOTICES *

JPO and NCIP are not responsible for any
damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

CLAIMS

[Claim(s)]

[Claim 1] The manufacture approach of the fullerene characterized by heating beforehand the temperature of the oxygen content gas which is combustion and/or the manufacture approach of fullerene which is made to carry out a pyrolysis and generates fullerene, and supplies a carbon content compound in a combustion furnace for a combustion reaction.

[Claim 2] The manufacture approach of the fullerene according to claim 1 characterized by being beyond the self-ignition temperature of the carbon content compound whose temperature of a preheating is a fuel.

[Claim 3] The manufacture approach of the fullerene according to claim 1 or 2 characterized by the oxygen content gas to supply containing inert gas.

[Claim 4] The manufacture approach of the fullerene according to claim 1 to 3 characterized by the oxygen content gas to supply being air.

[Claim 5] The manufacture approach of the fullerene according to claim 1 to 4 characterized by the pressure in a fission reactor being under atmospheric pressure.

[Claim 6] The manufacture approach of the fullerene characterized by being combustion and/or the manufacture approach of fullerene which is made to carry out a pyrolysis and generates fullerene about a carbon content compound, and a combustion condition being elevated-temperature air combustion into a combustion furnace.

[Claim 7] The manufacture approach of fullerene according to claim 1 to 6 that average combustion temperature is 1000 degrees C or more.

[Claim 8] The manufacture approach of fullerene according to claim 1 to 7 that the pressure in a fission reactor is under atmospheric pressure.

[Claim 9] The manufacturing installation of the fullerene which are combustion and/or the manufacture approach of fullerene which is made to carry out a pyrolysis and generates fullerene about a carbon content compound, and are characterized by for a fuel feed hopper and oxygen content gas supply opening separating distance independently respectively, and carrying out opening to the same fission reactor side into a combustion furnace.

[Claim 10] Equipment according to claim 9 which has a fuel feed hopper further in oxygen content gas supply inner mouth.

[Claim 11] Equipment according to claim 9 or 10 with the un-circular configuration of oxygen content gas supply opening which carried out opening into the fission reactor.

[Translation done.]

*** NOTICES ***

JPO and NCIP are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

DETAILED DESCRIPTION

[Detailed Description of the Invention]**[0001]**

[Field of the Invention] This invention relates to the manufacture approach of fullerene, and a manufacturing installation.

[0002]

[Description of the Prior Art] Fullerene (it may only be hereafter called fullerene) is the generic names of the third carbon allotrope which ranks second to a diamond and a graphite, and it is the carbon molecule of the shape of hollow husks closed in the network of five membered-rings and six membered-rings as represented in C₆₀, C₇₀, etc. Although it is comparatively that existence of fullerene was finally checked and it is a comparatively new carbon material, it is admitted that the special molecular structure, therefore specific physical property are shown, for example, innovative application development is being quickly developed over the wide range following fields.

(1) Application to a superhard ingredient : since purification of the artificial diamond which has a fine crystal grain child by using fullerene as a precursor is possible, use to an abrasion resistance material with added value is expected.

(2) Application to drugs : research as an application of an anticancer agent, an acquired immunodeficiency syndrome, osteoporosis and the Alzheimer remedy, a contrast medium, a stent ingredient, etc. is advanced by using C₆₀ derivative and an optical device.

(3) Application to a superconducting material : if metallic potassium is doped to a fullerene thin film, it is discovered that a superconducting material with a high transition temperature called 18K can be made, and since various, attract attention.

(4) Application to semi-conductor manufacture : it uses that resist structure is further strengthened with mixing C₆₀ with a resist, and the application to next-generation semi-conductor manufacture is expected.

[0003] Also in the fullerene of various carbon numbers, C₆₀ and C₇₀ are comparatively easy to compound, and it is expected that future need so also increases explosively. The approach shown below as the manufacture approach of fullerene learned now is mentioned.

(1) Laser vacuum deposition : how to irradiate the pulse laser of a high energy consistency at the carbon target placed into rare gas, and compound by evaporation of a carbon atom. The quartz tube with which rare gas flows is placed into an electric furnace, and a graphite sample is placed into the quartz tube. If laser is irradiated and is evaporated in a graphite sample from the upstream of the flow of gas, the soot containing fullerene, such as C₆₀ and C₇₀, will adhere to the wall of the quartz tube with which near the electric furnace outlet got cold. The evaporation per shot is slight and unsuitable for extensive manufacture.

(2) Resistance heating method : the approach to which carry out energization heating and a graphite rod is made to sublimate in the container of the vacuum filled with gaseous helium. Unsuitable for extensive manufacture, since the electric resistance loss in a circuit is large.

(3) Arc discharge method : the approach to which the carbon of a lifting and an anode plate is made to

sublimate arc discharge in the condition of having contacted two graphite electrodes lightly in the gaseous helium in dozens kPa(s), or having detached about 1-2mm. It is used for extensive manufacture on a current works scale.

(4) Radio frequency heating method : how to heat and evaporate a sink and this in an eddy current by RF induction at raw material graphite instead of using resistance heating and arc discharge.

(5) Combustion method : the approach of carrying out the incomplete combustion of the hydrocarbon raw materials, such as benzene, in the mixed gas of inert gas, such as helium, and oxygen. It is observed as the mass-producing method for being usable to liquid fuel etc., and the point that a manufacturing installation is simple, and opposing an arc synthesis method in the soot (fullerene etc.) reproduced in that several% of a benzene fuel serves as soot, and the about 10% becomes fullerene although manufacture effectiveness is not good.

(6) Naphthalene thermal decomposition method : the approach of carrying out the pyrolysis of the naphthalene at about 1000 degrees C.

[0004] Thus, although the synthesis method of various fullerene by current is proposed, the method of manufacturing fullerene in large quantities cheaply by any approach until now is not established. A combustion method is considered one of these approaches of the cheapest and efficient manufacture approach, and the manufacture approach of the fullerene by burning a carbon inclusion in a flame in the Patent Publication Heisei No. 507879 [six to] official report, and collecting condensates in it is indicated. Although fullerene is contained in soot-like matter, such as fullerene, and it is generated, it has been a big technical problem how the rate that fullerene is contained in this soot-like matter is raised.

[0005] In order to raise the yield of fullerene in the above and the Patent Publication Heisei No. 507879 [six to] official report, the approach of supplying energy further is stated to the flame from the external energy source as raising flame temperature and its means. As a desirable energy source, electric resistance heating of an input style, microwave heating, discharge heating, and counterflow heating are mentioned.

[0006] Moreover, generally, manufacture of fullerene is performed under reduced pressure and a diluent may be introduced all over a reaction field. It is known whenever [these reduced pressure] that diluent concentration will affect the yield of the above-mentioned fullerene. In the above and the Patent Publication Heisei No. 507879 [six to] official report, pure oxygen is used as an oxidizer for a combustion reaction, and the argon is used as a diluent. This is considered to be effective in gathering the yield of fullerene.

[0007] However, the amount of the oxygen needed for combustion also becomes extensive, and pure oxygen becomes a special oxygen supply facility is required and expensive [the manufacturing cost of fullerene] as a result, when a bomb or supply equipment of dedication etc. tends to be required and it is going to manufacture fullerene on a scale of industry especially. So, it has not resulted in utilization for the reasons of the linear velocity which the volume increases at the time of the actuation under that combustion temperature becomes low since there are many rates of that a flame is not stabilized compared with pure oxygen since the oxygen density is low although it can guess easily use air as an oxidizer of combustion in order to reduce a manufacturing cost in a combustion method, or nitrogen, especially reduced pressure, and passes a nozzle become quick.

[0008] The so-called elevated-temperature air combustion method is known as a combustion method which brings average thermal flux close to the maximum thermal flux with low speed heat release velocity fully oxidation exothermic reaction compared with the usual combustion, and controls NOx in a common industrial use heating furnace on the other hand about the combustion method itself. As another combustion method which controls NOx in an industrial use heating furnace, the charge of furnace internal combustion injecting [directly] method is learned. It is the approach that a combustion air and a fuel are injected in a furnace from the independent nozzle in detail, surrounding combustion gas is attracted according to the self-recirculation-of-exhaust-gas effectiveness by the jet energy, and reduction of the oxygen density of a combustion air and the fall of the flame temperature at the time of combustion can also be hung down.

[0009] Moreover, the combustion method which combined the above-mentioned elevated-temperature air combustion method and the injecting [directly]-in furnace method is also known. However, there is no publication about the manufacture approach of fullerene and equipment in any official report. Since fullerene is various as the exotic material which bears the next generation, and new materials, it is observed, and development of the technique of manufacturing fullerene cheaply and easily in large quantities is desired.

[0010]

[Problem(s) to be Solved by the Invention] This invention is made in view of a situation which was mentioned above, combustion all over a combustion furnace is considered as uniform combustion in manufacture of the fullerene by the combustion method, and it aims at offering the approach of manufacturing fullerene cheaply and easily in large quantities.

[0011]

[Means for Solving the Problem] this invention persons acquired knowledge that fullerene is stably generable by carrying out the preheating of the temperature of the oxidizer supplied to combustion beyond ordinary temperature, and making the pressure in a combustion furnace reduced pressure, as a result of examining various optimal combustion methods which can manufacture fullerene in large quantities and cheaply.

[0012] Since elevated-temperature-izing of combustion temperature and stabilization of a flame were easily attained by furthermore heating the temperature of an oxidizer beforehand beyond ordinary temperature, knowledge [that ** can also be diluted with rare gas, such as an argon, not using pure oxygen as an oxidizer] was also acquired. since [moreover,] the oxygen density of an oxidizer is lowered by this approach -- expensive gas like pure oxygen -- not using -- **, for example, air, -- like -- acquisition -- knowledge [that fullerene is generable with an easy oxidizer] was also acquired.

[0013] Generally the yield of the fullerene in a combustion method becomes so high that combustion flame temperature is high. As an approach of raising flame temperature, although addition of oxygen is known, since pure oxygen is used as an oxidizer in manufacture of fullerene, addition of the oxygen beyond it is impossible. Here, flame temperature can be easily raised by heating beforehand the temperature of the oxidizer supplied in a furnace.

[0014] Furthermore, if this temperature is raised to beyond the self-ignition temperature of a fuel hydrocarbon, the temperature distribution in the fission reactor in combustion can become homogeneity more, and can generate fullerene more efficiently. Moreover, since a combustion reaction is stable by this approach, even if it reduces an oxygen density, fullerene is stably generable [a diluent is added to pure oxygen, and].

[0015] Furthermore, air can be used as an oxidizer by this approach. Thereby, the cost in fullerene manufacture can be reduced sharply. Since nitrogen is contained in air when using air, NOx is contained in exhaust gas and the concentration becomes so high that preheat temperature of air is made high. Although the so-called injecting [directly]-in furnace method which separates distance and introduces an oxidizer and a fuel independently, respectively in the approach of reducing the oxygen density of the air to supply, the so-called elevated-temperature air combustion method, or a furnace is learned in order to control this NOx concentration rise, fullerene can be stably manufactured using cheap air by applying these approaches to the fullerene manufacture approach, and making it the bottom of the pressure reduced pressure in a fission reactor.

[0016]

[Embodiment of the Invention] First, the manufacturing installation and approach of fullerene concerning this invention are explained. The whole example outline sectional view of the fullerene manufacturing installation which drawing 1 requires for this invention, and drawing 2 are the arrangement explanatory views of the nozzle for oxidation gas installation, and a fuel installation nozzle. Generally, oxygen content gas supply opening (3) to oxygen content gas is supplied for a fuel hydrocarbon from a fuel feed hopper (2), and a combustion gas style hot by burning these is generated toward the lower stream of a river of a fission reactor. As oxygen content gas, the gas which mixed non-flammable gas, such as argon gas, at a rate of arbitration can be used for air, oxygen gas, or these, from a

viewpoint of the yield of fullerene, oxygen gas is desirable and air is desirable from a viewpoint of the ease of carrying out of acquisition of oxygen content gas etc. In order to raise especially combustion temperature, before being supplied in a furnace, the preheating of these oxygen content gas is carried out. As the approach of a preheating, what kind of well-known approaches, such as heat exchange with the combustion gas which used the heat exchanger, and the so-called regeneration burner, may be used. With [the temperature of this preheating] ordinary temperature [beyond], what kind of temperature is sufficient, but in order to gather the yield of fullerene, the high temperature is more desirable as much as possible. It is desirable more preferably that it is beyond the self-ignition temperature of a fuel. In this case, although the highest preheat temperature of an oxidizer is decided by heat-resistant temperature of the metal to be used by the general metal heat exchanger, when the heat-resistant metal generally known is used, about a maximum of 600 degrees C is a limitation. Therefore, when it is going to carry out a preheating beyond self-ignition temperature, it may be required to use a burner with special structures, such as a REJIENERESHON burner.

[0017] It is very good in the approach of making the self-recycling style of a combustion gas forming in a furnace by making the supply rate of flow into the furnace of an oxidizer into a high speed as an approach of heating beforehand the temperature of the oxidizer contributed to a combustion reaction in a furnace. In this case, the rate of flow of the oxygen content gas supplied into a furnace has 5 or more desirable m/s, and they are 10 or more m/s more preferably. Moreover, before it makes the configuration of an oxidizer feed hopper into a rectangle or oxygen content gas contacts a fuel at least in a furnace with devising the location of the intersection of fuel gas and oxidizer gas, the exhaust gas in a furnace can be involved in and temperature of oxygen content gas can also be carried out to beyond the self-ignition temperature of a fuel. In this case, in order to also dilute the oxygen density in oxygen content gas further and to fall, combustion in a furnace can also be made into the so-called elevated-temperature air combustion condition. In the state of such combustion, the temperature in a furnace is more uniform, and since it serves as an elevated temperature, it can raise the yield of fullerene more.

[0018] In order to maintain the stable elevated-temperature air combustion, by preparing a fuel feed hopper in oxygen content gas supply opening so that it may mention later, and usually burning a part of oxygen content gas, the temperature of oxygen content gas may be raised and an oxygen density may be reduced. As a fuel hydrocarbon and coal-for-coke-making-ized hydrogen, coal system liquid fuel, such as petroleum system liquid fuel, such as fuel gas, such as hydrogen, a carbon monoxide, natural gas, and petroleum gas, and a fuel oil, and creosote oil, can be used. Especially, it is desirable to use the aromatic series system hydrocarbon which refined these, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so good that its purity is close to 100% in case the purity of a raw material uses an aromatic series system hydrocarbon especially.

[0019] Moreover, in order to gather the yield of fullerene, it is desirable to also dilute combustion using rare gas etc. In order to make the combustion condition in a furnace into the so-called elevated-temperature air combustion condition, it is very good in what kind of well-known approach. In the fullerene manufacturing installation shown in drawing 1, opening of a fuel feed hopper (2) and the oxygen content gas supply opening (3) is respectively carried out to the same fission reactor side independently. The configuration of each feed hopper which is carrying out opening into the fission reactor may be arbitrary, and may be the indeterminate form of the shape of a polygon, such as the shape of an approximate circle form and an ellipse, and the shape of a trigonum and a rectangular head, a gourd mold, etc. According to this invention persons' knowledge, in the configuration in which a circular twist also has a major axis and a minor axis like the diameter of an ellipse, or a rectangle, heating of oxygen content gas and the rate of dilution speed up more. Therefore, as a fuel feed hopper (2), the shape of an ellipse and an approximate circle form are desirable, as oxygen content gas supply opening (3), the shape of a rectangle, such as the shape of a slit, is desirable, and it is desirable especially to combine these.

[0020] If opening of the arrangement of a fuel feed hopper (2) and oxygen content gas supply opening (3) is respectively carried out to the same fission reactor side independently, it is arbitrary. Although

various arrangement as shown in drawing 2 (A) - (E) can be taken according to furnace design conditions, such as a load of a fuel, and a burner number, if each feed hopper is especially arranged by turns to a hoop direction like drawing 2 (D) on the identitas which makes the core of the shaft-orientations cross section of a fission reactor the center of a circle, or a concentric circle periphery, since a furnace internal combustion glow condition becomes more uniform, it will be desirable. In this case, when the configuration of an oxygen gas feed hopper (6) has a major axis and a minor axis, it is desirable to arrange so that the straight line prolonged from the major axis may pass along the core of a circle (refer to drawing 2 (E)). Moreover, even if the open end is on the wall surface in a fission reactor, and an abbreviation same flat surface, although any feed hopper may be projected, an abbreviation same flat-surface top is preferably good [the feed hopper].

[0021] Although the fuel style and the oxygen content gas stream which are supplied in a fission reactor from a fuel feed hopper (2) and oxygen content gas supply opening (3) may be supplied from each open end at an angle of arbitration to the furnace wall side where each feed hopper is arranged, it is desirable to supply so that the fuel and/or oxygen content gas which are supplied may be further spread in the shape of an approximately concentric circle from an open end to Nagare's core so that it may become an abbreviation perpendicular preferably.

[0022] Moreover, in the range which satisfies the requirements for this invention, a fuel feed hopper (2) may be further prepared, for example into oxygen content gas supply opening (3). When the temperature of this in furnaces, such as the time of starting of a furnace, is low temperature, Or by supplying a fuel from the fuel feed hopper (2) installed into this oxygen content gas supply opening (3) and changing a combustion condition locally controlling the combustion temperature in a furnace, the physical properties of fullerene, etc., even if it is an elevated temperature It is because the combustion condition in a furnace can be controlled and more stable operation can be performed.

[0023] What is necessary is just to adjust the rate of flow of the oxygen content gas stream supplied in a fission reactor, and a fuel style according to the temperature change in a fission reactor etc. while choosing suitably. In the fullerene manufacturing installation shown in drawing 1 , a fuel feed hopper (4) and oxygen content gas supply opening (5) are further prepared in the furnace center section. The combustion temperature in a furnace is important and it is desirable to make 1400 degrees C or more 1800 more degrees C or more especially into 2000 degrees C or more also in at least 1000 degrees C or more. As for furnace internal pressure, it is desirable that it is under atmospheric pressure, and the more desirable range is 10 - 300torr.

[0024] The 0-40-mol range of the diluent concentration of a fuel is % substantially, and the diluent concentration of oxygen content gas can be adjusted to arbitration in the range of a mol 0 to 90%.

[0025]

[Effect of the Invention] By the manufacture approach of this invention, the extensive and cheap manufacture approach of fullerene by the economical combustion method can be offered.

[Translation done.]

*** NOTICES ***

**JPO and NCIP are not responsible for any
damages caused by the use of this translation.**

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

TECHNICAL FIELD

[Field of the Invention] This invention relates to the manufacture approach of fullerene, and a manufacturing installation.

[Translation done.]

*** NOTICES ***

JPO and NCIP are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

PRIOR ART

[Description of the Prior Art] Fullerene (it may only be hereafter called fullerene) is the generic names of the third carbon allotrope which ranks second to a diamond and a graphite, and it is the carbon molecule of the shape of hollow husks closed in the network of five membered-rings and six membered-rings as represented in C₆₀, C₇₀, etc. Although it is comparatively that existence of fullerene was finally checked and it is a comparatively new carbon material, it is admitted that the special molecular structure, therefore specific physical property are shown, for example, innovative application development is being quickly developed over the wide range following fields.

- (1) Application to a superhard ingredient : since purification of the artificial diamond which has a fine crystal grain child by using fullerene as a precursor is possible, use to an abrasion resistance material with added value is expected.
- (2) Application to drugs : research as an application of an anticancer agent, an acquired immunodeficiency syndrome, osteoporosis and the Alzheimer remedy, a contrast medium, a stent ingredient, etc. is advanced by using C₆₀ derivative and an optical device.
- (3) Application to a superconducting material : if metallic potassium is doped to a fullerene thin film, it is discovered that a superconducting material with a high transition temperature called 18K can be made, and since various, attract attention.
- (4) Application to semi-conductor manufacture : it uses that resist structure is further strengthened with mixing C₆₀ with a resist, and the application to next-generation semi-conductor manufacture is expected.

[0003] Also in the fullerene of various carbon numbers, C₆₀ and C₇₀ are comparatively easy to compound, and it is expected that future need so also increases explosively. The approach shown below as the manufacture approach of fullerene learned now is mentioned.

- (1) Laser vacuum deposition : how to irradiate the pulse laser of a high energy consistency at the carbon target placed into rare gas, and compound by evaporation of a carbon atom. The quartz tube with which rare gas flows is placed into an electric furnace, and a graphite sample is placed into the quartz tube. If laser is irradiated and is evaporated in a graphite sample from the upstream of the flow of gas, the soot containing fullerene, such as C₆₀ and C₇₀, will adhere to the wall of the quartz tube with which near the electric furnace outlet got cold. The evaporation per shot is slight and unsuitable for extensive manufacture.
- (2) Resistance heating method : the approach to which carry out energization heating and a graphite rod is made to sublimate in the container of the vacuum filled with gaseous helium. Unsuitable for extensive manufacture, since the electric resistance loss in a circuit is large.
- (3) Arc discharge method : the approach to which the carbon of a lifting and an anode plate is made to sublimate arc discharge in the condition of having contacted two graphite electrodes lightly in the gaseous helium in dozens kPa(s), or having detached about 1-2mm. It is used for extensive manufacture on a current works scale.
- (4) Radio frequency heating method : how to heat and evaporate a sink and this in an eddy current by RF induction at raw material graphite instead of using resistance heating and arc discharge.

(5) Combustion method : the approach of carrying out the incomplete combustion of the hydrocarbon raw materials, such as benzene, in the mixed gas of inert gas, such as helium, and oxygen. It is observed as the mass-producing method for being usable to liquid fuel etc., and the point that a manufacturing installation is simple, and opposing an arc synthesis method in the soot (fullerene etc.) reproduced in that several% of a benzene fuel serves as soot, and the about 10% becomes fullerene although manufacture effectiveness is not good.

(6) Naphthalene thermal decomposition method : the approach of carrying out the pyrolysis of the naphthalene at about 1000 degrees C.

[0004] Thus, although the synthesis method of various fullerene by current is proposed, the method of manufacturing fullerene in large quantities cheaply by any approach until now is not established. A combustion method is considered one of these approaches of the cheapest and efficient manufacture approach, and the manufacture approach of the fullerene by burning a carbon inclusion in a flame in the Patent Publication Heisei No. 507879 [six to] official report, and collecting condensates in it is indicated. Although fullerene is contained in soot-like matter, such as fullerene, and it is generated, it has been a big technical problem how the rate that fullerene is contained in this soot-like matter is raised.

[0005] In order to raise the yield of fullerene in the above and the Patent Publication Heisei No. 507879 [six to] official report, the approach of supplying energy further is stated to the flame from the external energy source as raising flame temperature and its means. As a desirable energy source, electric resistance heating of an input style, microwave heating, discharge heating, and counterflow heating are mentioned.

[0006] Moreover, generally, manufacture of fullerene is performed under reduced pressure and a diluent may be introduced all over a reaction field. It is known whenever [these reduced pressure] that diluent concentration will affect the yield of the above-mentioned fullerene. In the above and the Patent Publication Heisei No. 507879 [six to] official report, pure oxygen is used as an oxidizer for a combustion reaction, and the argon is used as a diluent. This is considered to be effective in gathering the yield of fullerene.

[0007] However, the amount of the oxygen needed for combustion also becomes extensive, and pure oxygen becomes a special oxygen supply facility is required and expensive [the manufacturing cost of fullerene] as a result, when a bomb or supply equipment of dedication etc. tends to be required and it is going to manufacture fullerene on a scale of industry especially. So, it has not resulted in utilization for the reasons of the linear velocity which the volume increases at the time of the actuation under that combustion temperature becomes low since there are many rates of that a flame is not stabilized compared with pure oxygen since the oxygen density is low although it can guess easily use air as an oxidizer of combustion in order to reduce a manufacturing cost in a combustion method, or nitrogen, especially reduced pressure, and passes a nozzle become quick.

[0008] The so-called elevated-temperature air combustion method is known as a combustion method which brings average thermal flux close to the maximum thermal flux with low speed heat release velocity fully oxidation exothermic reaction compared with the usual combustion, and controls NOx in a common industrial use heating furnace on the other hand about the combustion method itself. As another combustion method which controls NOx in an industrial use heating furnace, the charge of furnace internal combustion injecting [directly] method is learned. It is the approach that a combustion air and a fuel are injected in a furnace from the independent nozzle in detail, surrounding combustion gas is attracted according to the self-recirculation-of-exhaust-gas effectiveness by the jet energy, and reduction of the oxygen density of a combustion air and the fall of the flame temperature at the time of combustion can also be hung down.

[0009] Moreover, the combustion method which combined the above-mentioned elevated-temperature air combustion method and the injecting [directly]-in furnace method is also known. However, there is no publication about the manufacture approach of fullerene and equipment in any official report. Since fullerene is various as the exotic material which bears the next generation, and new materials, it is observed, and development of the technique of manufacturing fullerene cheaply and easily in large

quantities is desired.

[Translation done.]

THIS PAGE BLANK (USPTO)

*** NOTICES ***

JPO and NCIPI are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

EFFECT OF THE INVENTION

[Effect of the Invention] By the manufacture approach of this invention, the extensive and cheap manufacture approach of fullerene by the economical combustion method can be offered.

[Translation done.]

*** NOTICES ***

JPO and NCIPI are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] This invention is made in view of a situation which was mentioned above, combustion all over a combustion furnace is considered as uniform combustion in manufacture of the fullerene by the combustion method, and it aims at offering the approach of manufacturing fullerene cheaply and easily in large quantities.

[Translation done.]

* NOTICES *

JPO and NCIP are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

MEANS

[Means for Solving the Problem] this invention persons acquired knowledge that fullerene is stably generable by carrying out the preheating of the temperature of the oxidizer supplied to combustion beyond ordinary temperature, and making the pressure in a combustion furnace reduced pressure, as a result of examining various optimal combustion methods which can manufacture fullerene in large quantities and cheaply.

[0012] Since elevated-temperature-izing of combustion temperature and stabilization of a flame were easily attained by furthermore heating the temperature of an oxidizer beforehand beyond ordinary temperature, knowledge [that ** can also be diluted with rare gas, such as an argon, not using pure oxygen as an oxidizer] was also acquired. since [moreover,] the oxygen density of an oxidizer is lowered by this approach -- expensive gas like pure oxygen -- not using -- **, for example, air, -- like -- acquisition -- knowledge [that fullerene is generable with an easy oxidizer] was also acquired.

[0013] Generally the yield of the fullerene in a combustion method becomes so high that combustion flame temperature is high. As an approach of raising flame temperature, although addition of oxygen is known, since pure oxygen is used as an oxidizer in manufacture of fullerene, addition of the oxygen beyond it is impossible. Here, flame temperature can be easily raised by heating beforehand the temperature of the oxidizer supplied in a furnace.

[0014] Furthermore, if this temperature is raised to beyond the self-ignition temperature of a fuel hydrocarbon, the temperature distribution in the fission reactor in combustion can become homogeneity more, and can generate fullerene more efficiently. Moreover, since a combustion reaction is stable by this approach, even if it reduces an oxygen density, fullerene is stably generable [a diluent is added to pure oxygen, and].

[0015] Furthermore, air can be used as an oxidizer by this approach. Thereby, the cost in fullerene manufacture can be reduced sharply. Since nitrogen is contained in air when using air, NOx is contained in exhaust gas and the concentration becomes so high that preheat temperature of air is made high. Although the so-called injecting [directly]-in furnace method which separates distance and introduces an oxidizer and a fuel independently, respectively in the approach of reducing the oxygen density of the air to supply, the so-called elevated-temperature air combustion method, or a furnace is learned in order to control this NOx concentration rise, fullerene can be stably manufactured using cheap air by applying these approaches to the fullerene manufacture approach, and making it the bottom of the pressure reduced pressure in a fission reactor.

[0016]

[Embodiment of the Invention] First, the manufacturing installation and approach of fullerene concerning this invention are explained. The whole example outline sectional view of the fullerene manufacturing installation which drawing 1 requires for this invention, and drawing 2 are the arrangement explanatory views of the nozzle for oxidation gas installation, and a fuel installation nozzle. Generally, oxygen content gas supply opening (3) to oxygen content gas is supplied for a fuel hydrocarbon from a fuel feed hopper (2), and a combustion gas style hot by burning these is generated toward the lower stream of a river of a fission reactor. As oxygen content gas, the gas which mixed non-

flammable gas, such as argon gas, at a rate of arbitration can be used for air, oxygen gas, or these, from a viewpoint of the yield of fullerene, oxygen gas is desirable and air is desirable from a viewpoint of the ease of carrying out of acquisition of oxygen content gas etc. In order to raise especially combustion temperature, before being supplied in a furnace, the preheating of these oxygen content gas is carried out. As the approach of a preheating, what kind of well-known approaches, such as heat exchange with the combustion gas which used the heat exchanger, and the so-called regeneration burner, may be used. With [the temperature of this preheating] ordinary temperature [beyond], what kind of temperature is sufficient, but in order to gather the yield of fullerene, the high temperature is more desirable as much as possible. It is desirable more preferably that it is beyond the self-ignition temperature of a fuel. In this case, although the highest preheat temperature of an oxidizer is decided by heat-resistant temperature of the metal to be used by the general metal heat exchanger, when the heat-resistant metal generally known is used, about a maximum of 600 degrees C is a limitation. Therefore, when it is going to carry out a preheating beyond self-ignition temperature, it may be required to use a burner with special structures, such as a REJIENERESHON burner.

[0017] It is very good in the approach of making the self-recycling style of a combustion gas forming in a furnace by making the supply rate of flow into the furnace of an oxidizer into a high speed as an approach of heating beforehand the temperature of the oxidizer contributed to a combustion reaction in a furnace. In this case, the rate of flow of the oxygen content gas supplied into a furnace has 5 or more desirable m/s, and they are 10 or more m/s more preferably. Moreover, before it makes the configuration of an oxidizer feed hopper into a rectangle or oxygen content gas contacts a fuel at least in a furnace with devising the location of the intersection of fuel gas and oxidizer gas, the exhaust gas in a furnace can be involved in and temperature of oxygen content gas can also be carried out to beyond the self-ignition temperature of a fuel. In this case, in order to also dilute the oxygen density in oxygen content gas further and to fall, combustion in a furnace can also be made into the so-called elevated-temperature air combustion condition. In the state of such combustion, the temperature in a furnace is more uniform, and since it serves as an elevated temperature, it can raise the yield of fullerene more.

[0018] In order to maintain the stable elevated-temperature air combustion, by preparing a fuel feed hopper in oxygen content gas supply opening so that it may mention later, and usually burning a part of oxygen content gas, the temperature of oxygen content gas may be raised and an oxygen density may be reduced. As a fuel hydrocarbon and coal-for-coke-making-ized hydrogen, coal system liquid fuel, such as petroleum system liquid fuel, such as fuel gas, such as hydrogen, a carbon monoxide, natural gas, and petroleum gas, and a fuel oil, and creosote oil, can be used. Especially, it is desirable to use the aromatic series system hydrocarbon which refined these, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so good that its purity is close to 100% in case the purity of a raw material uses an aromatic series system hydrocarbon especially.

[0019] Moreover, in order to gather the yield of fullerene, it is desirable to also dilute combustion using rare gas etc. In order to make the combustion condition in a furnace into the so-called elevated-temperature air combustion condition, it is very good in what kind of well-known approach. In the fullerene manufacturing installation shown in drawing 1, opening of a fuel feed hopper (2) and the oxygen content gas supply opening (3) is respectively carried out to the same fission reactor side independently. The configuration of each feed hopper which is carrying out opening into the fission reactor may be arbitrary, and may be the indeterminate form of the shape of a polygon, such as the shape of an approximate circle form and an ellipse, and the shape of a trigonum and a rectangular head, a gourd mold, etc. According to this invention persons' knowledge, in the configuration in which a circular twist also has a major axis and a minor axis like the diameter of an ellipse, or a rectangle, heating of oxygen content gas and the rate of dilution speed up more. Therefore, as a fuel feed hopper (2), the shape of an ellipse and an approximate circle form are desirable, as oxygen content gas supply opening (3), the shape of a rectangle, such as the shape of a slit, is desirable, and it is desirable especially to combine these.

[0020] If opening of the arrangement of a fuel feed hopper (2) and oxygen content gas supply opening

(3) is respectively carried out to the same fission reactor side independently, it is arbitrary. Although various arrangement as shown in drawing 2 (A) - (E) can be taken according to furnace design conditions, such as a load of a fuel, and a burner number, if each feed hopper is especially arranged by turns to a hoop direction like drawing 2 (D) on the identitas which makes the core of the shaft-orientations cross section of a fission reactor the center of a circle, or a concentric circle periphery, since a furnace internal combustion glow condition becomes more uniform, it will be desirable. In this case, when the configuration of an oxygen gas feed hopper (6) has a major axis and a minor axis, it is desirable to arrange so that the straight line prolonged from the major axis may pass along the core of a circle (refer to drawing 2 (E)). Moreover, even if the open end is on the wall surface in a fission reactor, and an abbreviation same flat surface, although any feed hopper may be projected, an abbreviation same flat-surface top is preferably good [the feed hopper].

[0021] Although the fuel style and the oxygen content gas stream which are supplied in a fission reactor from a fuel feed hopper (2) and oxygen content gas supply opening (3) may be supplied from each open end at an angle of arbitration to the furnace wall side where each feed hopper is arranged, it is desirable to supply so that the fuel and/or oxygen content gas which are supplied may be further spread in the shape of an approximately concentric circle from an open end to Nagare's core so that it may become an abbreviation perpendicular preferably.

[0022] Moreover, in the range which satisfies the requirements for this invention, a fuel feed hopper (2) may be further prepared, for example into oxygen content gas supply opening (3). When the temperature of this in furnaces, such as the time of starting of a furnace, is low temperature, Or by supplying a fuel from the fuel feed hopper (2) installed into this oxygen content gas supply opening (3) and changing a combustion condition locally controlling the combustion temperature in a furnace, the physical properties of fullerene, etc., even if it is an elevated temperature It is because the combustion condition in a furnace can be controlled and more stable operation can be performed.

[0023] What is necessary is just to adjust the rate of flow of the oxygen content gas stream supplied in a fission reactor, and a fuel style according to the temperature change in a fission reactor etc. while choosing suitably. In the fullerene manufacturing installation shown in drawing 1 , a fuel feed hopper (4) and oxygen content gas supply opening (5) are further prepared in the furnace center section. The combustion temperature in a furnace is important and it is desirable to make 1400 degrees C or more 1800 more degrees C or more especially into 2000 degrees C or more also in at least 1000 degrees C or more. As for furnace internal pressure, it is desirable that it is under atmospheric pressure, and the more desirable range is 10 - 300torr.

[0024] The 0-40-mol range of the diluent concentration of a fuel is % substantially, and the diluent concentration of oxygen content gas can be adjusted to arbitration in the range of a mol 0 to 90%.

[Translation done.]

*** NOTICES ***

JPO and NCIPI are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] Drawing 1 is the explanatory view of the whole example outline sectional view of the fullerene manufacturing installation concerning this invention and the nozzle for oxidation gas installation and a fuel installation nozzle, and the example of a raw material installation nozzle configuration.

[Drawing 2] Drawing 2 is the arrangement explanatory view of the nozzle for oxidation gas installation, a fuel installation nozzle, and a raw material installation nozzle.

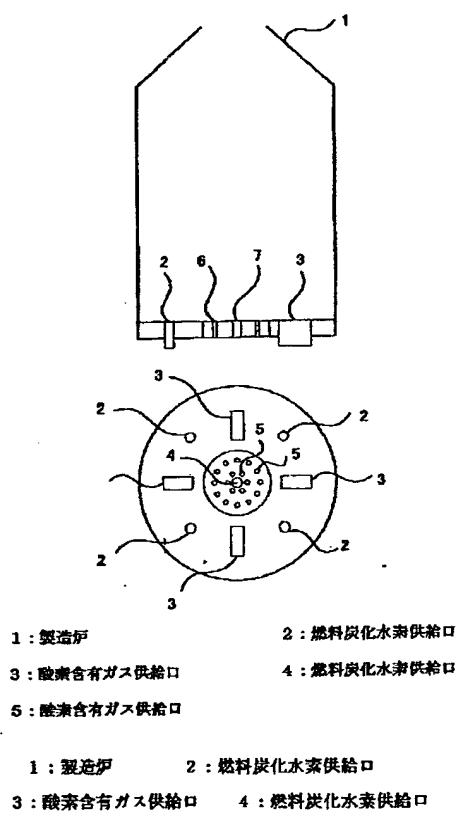
[Translation done.]

* NOTICES *

JPO and NCIPI are not responsible for any damages caused by the use of this translation.

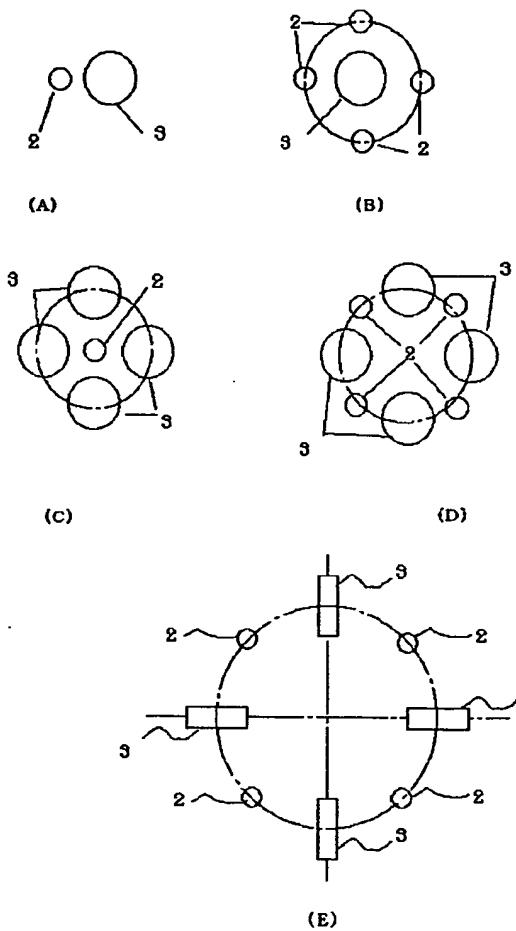
1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

DRAWINGS



[Drawing 1]

[Drawing 2]



[Translation done.]

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- BLACK BORDERS**
- IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**
- FADED TEXT OR DRAWING**
- BLURRED OR ILLEGIBLE TEXT OR DRAWING**
- SKEWED/SLANTED IMAGES**
- COLOR OR BLACK AND WHITE PHOTOGRAPHS**
- GRAY SCALE DOCUMENTS**
- LINES OR MARKS ON ORIGINAL DOCUMENT**
- REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**
- OTHER:** _____

**IMAGES ARE BEST AVAILABLE COPY.
As rescanning these documents will not correct the image
problems checked, please do not report these problems to
the IFW Image Problem Mailbox.**